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(54) **Additive for smokers tobacco**

(57) An additive for smokers tobacco, by which dangerous substances in tobacco smoke, such as Benzo(a)pyrenes, other polycyclic aromatic hydrocarbons, N-Nitrosamine and carbon monoxide, are reduced, and which consists of carbamide and/or amides of organic acids.

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SPECIFICATION

Additive for smokers tobacco

The invention relates to an additive for smokers tobacco, for reducing dangerous substances in the tobacco smoke.

Important dangerous substances in tobacco smoke, according to present knowledge, are benzo(a)pyrene (BaP) and other polycyclic aromatic hydrocarbons (PaH) and the N-nitrosamines. The carcinogenic action of these substances seems to be certain. As co-carcinogenic compounds, which synergetically cooperate with the said carcinogens, and especially with BaP, the phenols come into consideration. A further dangerous substance in tobacco smoke is carbon monoxide (CO) which combines with the haemoglobin of the blood.

According to the state of the art, a variety of methods are known for reducing dangerous materials in tobacco smoke, which at least partly are in practical use. There has been successful development of new tobacco varieties for reducing the condensate proportion in the tobacco smoke. By a special fertiliser treatment of the tobacco plant, a lowering of the proportion of condensate and N-nitrosamine can be achieved. These measures lead to the presently widely used low nicotine tobacco varieties, which compared with others produce less condensate and thus also less of the dangerous substances on smoking. These biological and agrarian procedures naturally have limits. For further reduction of dangerous substances supplementary measures must therefore be taken.

More particularly, the use of various filters, or extending or diluting the tobacco with various substances, and the use of porous, air permeable cigarette papers, comes into consideration. In this manner, a further lowering of the tobacco smoke condensate is achieved. However, these measures are frequently accompanied by a change in taste and a lowering of the exciting action of the tobacco smoke. Consequently, such cigarettes are not accepted by a large proportion of smokers.

An alternative or supplementary procedure is the treating of the tobacco with additives, for example nitrates, chlorates, citric acid and deuterium oxide as well as heavy and noble metals or their combinations. These methods have mainly the object of reducing the proportion of PaH more particularly of BaP in the tobacco smoke.

All previously proposed methods for treating the tobacco with additives, as already mentioned, have either heavy disadvantages or can not withstand critical testing with regard to their action. It has been found that tobacco treatment with citric acid, for examples, does not lead to a positive result. Treatment with deuterium oxide serves simply for simulating a lowering of the BaP content in the tobacco smoke, since deuterium oxide indicates weaker in conventional analysis methods. Nitrates are customarily used as treatment means for tobacco, but are questionable because of the danger of N-nitrosamine formation with the pyrolysis and are prohibited in some countries. Tobacco treatment with noble metals and their compounds leads only to a slight reduction of the tobacco smoke condensate without an additional specific lowering of the content of BaP and other PAH. The high price of these substances used for the treatment makes an economic application of the method difficult. Some of the substances investigated for possible use for tobacco treatment are toxic and/or form volatile toxic compounds on smoking of the tobacco. This is for example the case with nickel and copper salts, other heavy metals and their compounds.

It is an object of the invention to provide additives for tobacco, by which dangerous materials in the tobacco smoke can be considerably reduced, and in which on smoking of tobacco products treated with the additive, more particularly the proportion of benzo(a)pyrene (BaP), other polycyclic aromatic hydrocarbons (PAH) and carbon monoxide (CO) are reduced as well as the formation of N-nitrosamines supressed, and which can be used in large scale production.

This object is solved by an additive which consists of carbamide and/or amides of organic acids.

The treatment of the tobacco is effected in such manner that it is steeped, sprayed or coated with an aqueous solution, an emulsion or a dispersion of one or more of the additives.

In the search for suitable non-toxic substances for tobacco treatment it has surprisingly been found that treatment of tobacco with carbamide, oxalic acid diamide and other amides of organic acids leads to a reduction of dangerous substances, which far exceeds all known treatments in this respect. The treatment more particularly causes a significant reduction of the BaP content as well as the carbon monoxide in the tobacco smoke. Such a reduction of the dangerous substances markedly reduces the health risk of the smoker, more particularly the strongly cancer liable heavy smoker, without such person having to drastically alter smoking habits.

In the series of additives according to the invention, carbamide is the most significant. Carbamide is a physiologically completely harmless substance. Nothing is known with regard to any toxic action of carbamide. As volatile pyrolysis products of carbamide, under the conditions which occur on smoking of tobacco, there are simply carbon dioxide (CO_2) and ammonia (NH_3). Biuret which possibly occurs as an intermediate product is converted at the combustion temperatures of a cigarette, in the ambience of the always present water, into carbon dioxide and ammonia. The ammonia which is formed, freely and completely combines with the substances contained in the tobacco or their pyrolysis products, so that on smoking tobacco treated with carbamide, even up to as much as five per cent carbamide by weight, practically no ammonia can be detected in the main smoke flow.

Tobacco treated with carbamide is essentially completely neutral in taste. Any minimal difference in taste can easily be disguised by an appropriate flavouring of the tobacco. Moreover, carbamide is unlimitedly durable on the tobacco without any change.

The application of the carbamide onto or in the

tobacco does not cause any problems, even on a large production scale, due to the water solubility of this compound. No special procedures are necessary within the scope of conventional tobacco treatment. Carbamide can for example be applied during the moistening of the tobacco leaves before cutting, or together with other additive materials during the subsequent handling of the tobacco. A carbamide proportion of less then ten percent by weight does not practically alter the consistency of the tobacco fibre. Due to the solubility of the carbamide, application of the desired amount of mixed-in carbamide, which as a rule amounts to a few percent, can be done precisely and without problems.

The application of oxalic acid diamide and other organic acid amides is associated with greater difficulty, owing to the essentially poorer water solubility of these substances. They can for example be finely ground and applied in the form of a dispersion or possibly in the form of an emulsion onto the tobacco.

The entire tobacco amount can be treated with the additive or only a portion thereof prepared. If only a partial amount is treated, the amount of additive based on the tobacco weight must be greater than with treatment of the entire amount.

The use of the additives according to the invention for smoking tobacco, for reducing the dangerous substances occurring on smoking thereof, is economic, carbamide more particularly being a cost favourable substance which is always available in sufficient quantities. The increase in price of a cigarette due to the treatment according to the invention, can be kept within totally acceptable limits.

The manner of operation, more particularly of carbamide, in reducing the dangerous materials in the tobacco smoke, can be considered as follows:

The PAH and thus also the BaP occur on the pyrolysis of the tobacco, in a number of phases. A preliminary phase of the PAH are molocules of individual electrons, the so-called radicals. The ammonia occurring on the pyrolysis of carbamide and the amino groups of the carbamide possibly react with such radicals and thereby impede the production of part of the PAH or of the BaP.

Moreover, treatment of the tobacco with carbamide also impedes a production of N-nitrosamine on smoking, at least partially. These N-nitrosamines can occur by reaction of secondary and tertiary amines, for example nicotine, with nitrous gases which are released by the thermal splitting of the nitrates present in the tobacco. Primary amino groups and ammonia react with formation of nitrogen. If by the addition of carbamide to the tobacco a very great excess of primary amino groups or ammonia is established, then it is to be expected that the nitrous gases preferentially react with these. The possibility of formation of N-nitrosamines is thus essentially reduced.

On the bases of this mechanism of action, substances the molocules of which contain amino groups, and organic and inorganic ammonium salts, can under certain circumstances in principle lead to the same action as carbamide.

The explanation given of the action of carbamide in reducing dangerous substances is based on the dominant radical theory for the generation of polycyclics, but is naturally not definitive. It has been found that the production of other polycyclics in similar orders of magnitude as benzo(a)pyrene also occurs. It is assumed that not only free ammonia, but also the amino groups of the non-split carbamide, contribute to the observable action.

The invention will be further described with reference to several examples, from which the action will be clear.

Example 1.

A small amount of a fine cut tobacco was prepared by spraying with an aqueous carbamide solution, with five percent carbamide by weight. Parallel thereto, a similar amount of the same tobacco was sprayed with the same amount of distilled water. After drying of both tobacco samples to about twelve percent water content by weight, several hundred filter cigarettes were made therefrom by means of a hand machine. After testing these cigarettes for tobacco weight and resistance to drawing, the cigarettes were conditioned according to DIN 10244 and 10240 and machine smoked. The raw condensate, carbon monoxide in the tobacco smoke and benzo(a)pyrene in the tobacco smoke condensate were determined. Determination of the benzo(a)pyrene was effected after separating the disturbing accompanying substances fluorimetrically by column and thin layer chromatography.

On evaluating the results of the test, it was established that the treatment of the tobacco with five percent carbamide by weight, led to a statistically highly significant reduction of:
the benzo(a)pyrene by about 43%
and the raw condensate by about 20%
and an apparent reduction of the carbon monoxide by about fiteen percent in the tobacco smoke.

Example 2

Two large samples of fine cut tobacco were prepared as in example 1. From this tobacco 30000 filterless cigarettes were manufactured by machine per experiment. These were further treated as described in Example 1. The experiment results with the thus prepared cigarettes showed the following statistically highly significant reductions:
Reduction of Benzo(a)pyrene about 32%
Reduction of Raw condensate about 5%
Reduction of Carbon monoxide about 11%
Especially notable in these examples is that the treatment of the tobacco led in both cases to a significant lowering of the BaP content in the tobacco smoke condensate.

Contrary to a pure filtration which only quantitatively reduces the condensate amount in the tobacco smoke, in the present case as well as the quantitative reduction of the condensate amount a specific reduction of the BaP content in the condensate is achieved. Since the nicotine content of a cigarette ie essentially proportional to its condensate amount, the treatment of the tobacco with carbamide allows a lowering of the nicotine content in excess proportion to the lowering of the dangerous substances, to

be expected. Such an action corresponds to the requirements of the smoker.

Example 3

The smoke from cigarettes manufactured from conventional tobacco of average weight was investigated in the Battelle-Institute, Frankfurt. Part of this tobacco was treated with 5% carbamide by weight, the other part was prepared in the same way with distilled water. The cigarettes were selected according to the appropriate DIN standard and smoked. In the condensate, the volatile N-nitrosamine : N-nitrosodimethyl amine (NDMA), N-nitroso-ethyl methyl amine (NEMA) and N-nitroso-pyrrolidine (NPY) were determined. These substances could not be shown in the tobacco itself. In the tobacco smoke condensate, a statistically highly significant lowering of the content of all three substances was established, caused by the carbamide. Individually, for these reductions, the following values were obtained.

– NDMA
about 34% (based on 1g of cigarette)
about 37% (based on 1g of raw condensate)
– NEMA
about 43% (based on 1g of cigarette)
about 41% (based on 1g of raw condensate)
– NPY
about 22% (based on 1g of cigarette)
about 24% (based on 1g of raw condensate)

The reduction of the NDMA content is of particular significance, since NDMA has the greatest carcinogenic activity of all nitrosamines. The effect found for the NEMA, having regard to the low absolute concentration of this substance, is certainly of subordinate significance.

The comparatively slight reduction of the NPY content is apparently due to the presence of N - nitroso - nornicotine (NNN) in the tobacco : NPY occurs on smoking of NNN - containing tobacco, partly through thermal splitting of the already present NNN. Since carbamide can not possibly destroy any already present N-nitroso-groups, but only partly inhibit their fresh production, a reduction of NNN and thus secondarily a reduction of NPY can only be expected for the amount additionally formed with the tobacco combustion, which – measured for the total quantity – gives a small reduction value. Determination of the NNN requires another analysis technique and was thus not provided within the scope of these tests.

The following test is an example for an additive of an amide or an organic acid.

Example 4

The procedure was as in Example 1, but instead of a carbamide solution an aqueous solution of oxalic acid mono ethyl ester amide was used.

The reduction of Benzo(a)pyrene amounted to about 21%

The lower effect may be seen to be because a lower amount of amide is present in the molecules of the additive than in the carbamide.

CLAIMS

1. An additive for smokers tobacco, for reduction of dangerous substances in tobacco smoke, consisting of carbamide and/or amides of organic acids.

2. An additive according to claim 1, consisting of one or more amides of aliphatic multiple carbonic acid.

3. An additive according to claim 2, consisting of oxalic acid diamide or oxalic acid monoethyl ester amide.

4. Method for the treatment of smokers tobacco with an additive according to claim 1, 2 or 3, characterised in that the tobacco is steeped, sprayed or coated with an aqueous solution, an emulsion or a dispersion of one or more of the additives.

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